

## TRANSMITTAL LETTER TO THE UNITED STATES

214751US0PCT

DESIGNATED/ELECTED OFFICE (DO/EO/US)

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

CONCERNING A FILING UNDER 35 U.S.C. 371

09/926282

INTERNATIONAL APPLICATION NO.

PCT/JP00/02319

INTERNATIONAL FILING DATE

10 April 2000

PRIORITY DATE CLAIMED

8 April 1999

TITLE OF INVENTION

PREFORM FOR COMPOSITE MATERIAL AND COMPOSITE MATERIAL

APPLICANT(S) FOR DO/EO/US

MITANI Kazutami et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
  - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☒ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
- ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is attached hereto.
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- ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
- ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
- ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
- ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

## Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.
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19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
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22. ☐ Certificate of Mailing by Express Mail
23. ☒ Other items or information:

Request for Consideration of Documents Cited in International Search Report

Notice of Priority

PCT/IB/304

PCT/IB/308


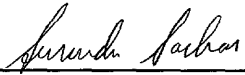
U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.492(a)(1)-(5)) : <b>091/926282</b>	INTERNATIONAL APPLICATION NO. <b>PCT/JP00/02319</b>	ATTORNEY'S DOCKET NUMBER <b>214751US0PCT</b>
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24. The following fees are submitted:.				<b>CALCULATIONS PTO USE ONLY</b>	
<b>BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5)) :</b>					
<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO . . . . .				<b>\$1040.00</b>	
<input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO . . . . .				<b>\$890.00</b>	
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO . . . . .				<b>\$740.00</b>	
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) . . . . .				<b>\$710.00</b>	
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) . . . . .				<b>\$100.00</b>	
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>\$890.00</b>	
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).				<b>\$0.00</b>	
<b>CLAIMS</b>	<b>NUMBER FILED</b>	<b>NUMBER EXTRA</b>	<b>RATE</b>		
Total claims	9 - 20 =	0	x \$18.00	<b>\$0.00</b>	
Independent claims	2 - 3 =	0	x \$84.00	<b>\$0.00</b>	
Multiple Dependent Claims (check if applicable).			<input checked="" type="checkbox"/>	<b>\$280.00</b>	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$1,170.00</b>	
Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.				<b>\$0.00</b>	
<b>SUBTOTAL =</b>				<b>\$1,170.00</b>	
Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).				<b>\$0.00</b>	
<b>TOTAL NATIONAL FEE =</b>				<b>\$1,170.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).			<input type="checkbox"/>	<b>\$0.00</b>	
<b>TOTAL FEES ENCLOSED =</b>				<b>\$1,170.00</b>	
				<b>Amount to be: refunded</b>	<b>\$</b>
				<b>charged</b>	<b>\$</b>

- a. ☒ A check in the amount of \$1,170.00 to cover the above fees is enclosed.
- b. ☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 15-0030 A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

<p align="center"><b>Surinder Sachar</b> <b>Registration No. 34,423</b></p> <p align="center"> <b>22850</b></p>	<p align="center"> SIGNATURE</p> <p align="center"><b>Norman F. Oblon</b> NAME</p> <p align="center"><b>24,618</b> REGISTRATION NUMBER</p> <p align="center"><u>Oct. 5 2001</u> DATE</p>
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FORM PTO-1390 (Modified)  
(REV 11-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

**TRANSMITTAL LETTER TO THE UNITED STATES**  
**DESIGNATED/ELECTED OFFICE (DO/EO/US)**  
**CONCERNING A FILING UNDER 35 U.S.C. 371**

214751US0PCT

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

**U9/926282**INTERNATIONAL APPLICATION NO.  
**PCT/JP00/02319**INTERNATIONAL FILING DATE  
**10 April 2000**PRIORITY DATE CLAIMED  
**8 April 1999**

TITLE OF INVENTION

**PREFORM FOR COMPOSITE MATERIAL AND COMPOSITE MATERIAL**

APPLICANT(S) FOR DO/EO/US

**MITANI Kazutami et al.**

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**Notice of Priority**  
**PCT/IB/304**  
**PCT/IB/308**

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SEND ALL CORRESPONDENCE TO:



**22850**  
**Surinder Sachar**  
**Registration No. 34,423**

*Surinder Sachar*

SIGNATURE

**Norman F. Oblon**

NAME

**24,618**

REGISTRATION NUMBER

*Oct. 5 2001*

DATE

DOCKET NO.: 214751US0PCT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN RE APPLICATION OF: MITANI Kazutami et al.

SERIAL NO.: NEW U.S. PCT APPLICATION

FILED: HERewith

INTERNATIONAL APPLICATION NO.: PCT/JP00/02319

INTERNATIONAL FILING DATE: April 10, 2000

FOR: PREFORM FOR COMPOSITE MATERIAL AND COMPOSITE MATERIAL

**REQUEST FOR PRIORITY UNDER 35 U.S.C. 119**  
**AND THE INTERNATIONAL CONVENTION**

Assistant Commissioner for Patents  
Washington, D.C. 20231


Sir:

In the matter of the above-identified application for patent, notice is hereby given that the applicant claims as priority:

<u>COUNTRY</u>	<u>APPLICATION NO</u>	<u>DAY/MONTH/YEAR</u>
Japan	11-101617	08 April 1999

Certified copies of the corresponding Convention application(s) were submitted to the International Bureau in PCT Application No. PCT/JP00/02319. Receipt of the certified copy(s) by the International Bureau in a timely manner under PCT Rule 17.1(a) has been acknowledged as evidenced by the attached PCT/IB/304.

Respectfully submitted,  
OBLON, SPIVAK, McCLELLAND,  
MAIER & NEUSTADT, P.C.



Norman F. Oblon  
Attorney of Record  
Registration No. 24,618  
Surinder Sachar  
Registration No. 34,423



22850

(703) 413-3000  
Fax No. (703) 413-2220  
(OSMMN 1/97)

DOCKET NO.: 214751US0PCT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN RE APPLICATION OF: MITANI Kazutami et al.

SERIAL NO.: NEW U.S. PCT APPLICATION

FILED: HERewith

INTERNATIONAL APPLICATION NO.: PCT/JP00/02319

INTERNATIONAL FILING DATE: April 10, 2000

FOR: PREFORM FOR COMPOSITE MATERIAL AND COMPOSITE MATERIAL

**REQUEST FOR CONSIDERATION OF DOCUMENTS**  
**CITED IN INTERNATIONAL SEARCH REPORT**

Assistant Commissioner for Patents

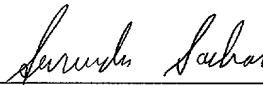
Washington, D.C. 20231

Sir:

In the matter of the above-identified application for patent, notice is hereby given that applicant(s) request that the Examiner consider the documents cited in the International Search Report according to MPEP §609 and so indicate by a statement in the first Office Action that the information has been considered. When the Form PCT/DO/EO/903 indicates both the search report and copies of the documents are present in the national stage file, there is no requirement for the applicant(s) to submit them (1156 O.G. 91 November 23, 1993).

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,  
MAIER & NEUSTADT, P.C.



Norman F. Oblon  
Attorney of Record  
Registration No. 24,618  
Surinder Sachar  
Registration No. 34,423



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(703) 413-3000

Fax No. (703) 413-2220

(OSMMN 1/97)

## DESCRIPTION

## PREFORM FOR COMPOSITE MATERIAL AND COMPOSITE MATERIAL

5

## Technical Field

This invention relates to a preform for a composite material which is preferably used for resin transfer molding, and to a fiber-reinforced composite material formed by molding the preform.

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## Background Art

As a preform material for resin transfer molding, conventionally, a random mat and a swirl mat are used. Furthermore, if high strength and elastic modulus are required, a laminated product formed of a long fiber fabric in which reinforcement fibers are arranged relatively in line and a material formed by stitching the laminated product so as to be engaged, are used. Recently, various preforms for manufacturing a braiding material having an involute shape, three-dimensional fabric, and the like, which have excellent properties, have been developed.

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In a composite material which is reinforced by using a laminated product formed of reinforcement fiber fabric such as a plain-woven fabric, satin woven fabric, and the like, since interlaminar debonding caused by an impact or the like considerably depends on toughness of a matrix resin, it is not easy to give interlaminar debonding resistance at a high level to a composite material formed by molding thermosetting resin using resin transfer molding for a matrix resin, wherein it is difficult to improve the toughness of the resin.

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A preform formed as a unit by stitching a laminated product by laminating fabric in the direction of thickness shows an effect of inhibiting the interlaminar debonding. However, on the other hand, a special sewing machine is required in order to stitch the three-dimensional laminated fabric, and there is a trade-off (inverse proportion) between improvement of the interlaminar debonding resistance by increasing stitch density in the direction of thickness and strength in the direction of the inside from surfaces of the laminated product.

In the braiding material and the three-dimensional fabric, reinforcement fiber can be arranged in the direction of thickness of the composite material, and therefore, a composite material having excellent properties can be obtained. However, if large structures are to be

manufactured, a machine for manufacturing preforms corresponding to the structures must be huge and the cost per unit of preform will be enormous.

Furthermore, in a composite material obtained by laminating and hardening a sheet type intermediate material (prepreg) made of reinforcement fiber impregnated with thermosetting resin, it is known that thermoplastic resin is provided between layers of a laminated product by a method such that a film, particles, fiber, non-woven fabric, and the like formed of thermoplastic resin are provided on surfaces of prepregs or between prepregs during a step of laminating prepregs in order to improve a property of interlaminar debonding resistance. However, in resin transfer molding in which a preform is formed with reinforcement fiber and subsequently, matrix resin is injected, it is necessary to ensure sufficient flow of the matrix resin when the matrix resin is injected. Therefore, the above method for improving interlaminar debonding resistance which is applied for prepregs cannot simply be applied to the resin transfer molding.

#### Disclosure of Invention

An object of the present invention is to provide a preform for a composite material and a composite material formed from the preform, wherein the composite material can be molded by resin transfer molding, having excellent strength and a property of interlaminar debonding resistance.

As a result of diligent research with regard to the preform for a composite material providing a composite material having excellent strength and excellent interlaminar debonding resistance, the present inventors have found that the above object can be achieved by using a laminated product formed of reinforcement fiber in which a thermoplastic resin layer having a space so as not to inhibit liquid resin from flowing therethrough, is provided between layers of the laminated product. The laminated product is preferably used as a preform for resin transfer molding.

That is, the present invention provides a preform for a fiber-reinforced composite material comprising a reinforcing material formed of reinforcement fiber forming layers as a laminated structure and a thermoplastic resin layer having space so as not to inhibit liquid resin from flowing therethrough, provided between the layers.

Furthermore, the present invention provides a fiber-reinforced composite material formed by molding the above preform for a composite material.



## Best Mode for Carrying Out the Invention

A reinforcing material made of reinforcement fiber used for a preform for a composite material is a fiber exhibiting excellent strength and/or elastic modulus such as carbon fiber, glass fiber, aramid fiber, and the like. In particular, carbon fiber is preferably used for excellent strength and elastic modulus.

The reinforcing material made of the above fiber is generally supplied as a fiber bundle (tow) composed of tens to tens of thousands of fibers. The fiber tows may be spread or be provided in the same direction to form a layer and the layers are put on each other so as to be laminated so that a fiber tow of the layer is provided in the different direction from another fiber tow to form a laminated structure. Furthermore, the reinforcing material may be formed into uniaxial, biaxial, or triaxial plane-woven fabric beforehand and at least one kind of the plane-woven fabrics is put on another to form a laminated structure. The uniaxial fabric is made by weaving fiber or fiber tows which are provided in a predetermined direction (longitudinal direction) on a planar surface and a low number of threads (weft) in lateral or oblique direction, or by bonding and engaging the fiber or fiber tows to a low number of threads in a lateral or oblique direction. The uniaxial fabric refers to a sheet composed of practically only fiber tows provided in the longitudinal direction.

A resin forming the thermoplastic resin layer which is provided between the layers composed of reinforcing material formed of the above reinforcement fiber, is not limited as long as the resin can be processed into porous films, fibers, reticulated structures, or knitted loops. The thermoplastic resins, which have excellent toughness, such as polyamides, polyether imides, polyether ether ketones, polyimides, and the like are preferably used, and furthermore, the resin is selected from the above thermoplastic resins depending on the environment for use required by the desired composite material or on a matrix resin used for the composite material.

The above thermoplastic resin layer provided between the layers composed of reinforcing material formed of the reinforcement fibers is required to provide space appropriately so as not to inhibit liquid resin from flowing therethrough. The phrase "so as not to inhibit liquid resin from flowing therethrough" means "the thermoplastic resin layer does not inhibit liquid matrix resin from flowing in the preform" when the liquid matrix resin is injected into the preform in order to mold a composite material by the resin transfer molding. The space may be provided suitably, unless resistance generated when fluid flows

in the thermoplastic resin layer is remarkably greater than resistance generated when fluid flows through the reinforcing material formed of the reinforcement fiber. A form of the thermoplastic resin layer as described above is selected from the group consisting of porous film, fiber, network structure, knitted loop, oriented fibers formed between layers of the reinforcing material formed of the reinforcement fiber, an aggregation of short fibers, an aggregation of particles, and the like.

The amount of the resin for forming the above thermoplastic resin layer which is provided between the layers composed of reinforcing material formed of the reinforcement fiber, is preferably set within  $1 \text{ g/m}^2$  to  $50 \text{ g/m}^2$  based on areal density per thermoplastic resin layer. If the amount of the resin is less than  $1 \text{ g/m}^2$ , fracture toughness, which prevents interlaminar debonding, is not sufficiently exhibited, and if the amount of the resin is larger than  $50 \text{ g/m}^2$ , the width between the layers of the reinforcing material becomes thicker and stress transfer is insufficient when the resin is used in the composite material.

If the thermoplastic resin layer provided between the layers composed of reinforcing material formed of the reinforcement fiber is composed of fibers of the thermoplastic resin, although a monofilament of the fiber is not limited in denier, 1 to 50 deniers (d) are preferably used. If the monofilament is finer than 1 d, the monofilament tends to be carried along by the flow of the liquid matrix resin during resin transfer molding, and sometimes the monofilaments break and are unevenly distributed. If the monofilament is thicker than 50 d, the area of an interface between the thermoplastic resin in the fiber and the matrix resin becomes smaller, and therefore interlaminar debonding resistance is not easily given.

The thermoplastic resin fiber provided between the layers formed of reinforcing material composed of the reinforcement fiber can be provided as monofilaments and also as fiber bundles (tows). If the fiber tow of thermoplastic resin is used, the fiber tow is spread or is cut and homogenously dispersed to be evenly provided between the layers composed of the reinforcing material formed of the reinforcement fiber. Furthermore, a row of provided fiber tows or spread fiber tows with remaining space between the fiber tows or a fabric made of the fiber tows can be used by providing it between the layers formed of reinforcing material composed of the reinforcement fiber. For example, the fiber tows formed of the thermoplastic resin are provided with spaces between one another on a reinforcing uniaxial fabric perpendicularly to the reinforcing direction of the uniaxial fabric formed of the reinforcement fiber.

A row of provided fiber tows or spread fiber tows with remaining space between the fiber tows or a fabric made of the fiber tow can be used by providing them between the layers formed of reinforcing material composed of the reinforcement fiber, and a ratio of the surface of the reinforcing material layer formed of the reinforcement fiber covered with the fiber bundle formed of the thermoplastic resin, is preferably over 20% in any square 1 cm on a side on the surface of the reinforcing material layer formed of reinforcement fiber.

Providing the above thermoplastic resin layer between the layers formed of reinforcing material composed of the reinforcement fiber is possible by, but is not limited to, forming a reinforcing material layer composed of the reinforcement fiber or a thermoplastic layer on the surface of an alternating laminate of reinforcing material layers composed of the reinforcement fiber and thermoplastic resin layers which have already been formed in the same manner, alternately laminating the reinforcing material layer composed of the reinforcement fiber and the thermoplastic resin layer which have been prepared respectively, or laminating sheets composed of the reinforcement fiber and the thermoplastic resin layer wherein the sheet is prepared by forming a thermoplastic resin layer on at least one surface of a reinforcing material sheet composed of the reinforcement fiber. If fiber or tape formed of thermoplastic resin is used as a base material forming a thermoplastic resin layer, fabrics formed by weaving the reinforcement fiber and the fiber or the tape of thermoplastic resin as the weft and warp are laminated.

The preform for composite material having a laminated structure according to the present invention is characterized in that the preform provides a composite material having excellent interlaminar debonding property by being molded by resin transfer molding. Moreover, stitching can be applied through the thickness of the reinforcing material layers formed of the reinforcement fiber.

As a matrix resin of the composite material used for the present invention, a thermosetting resin such as unsaturated polyester resin, vinyl ester resin, epoxy resin, bismaleimide resin, isocyanate resin, and the like is used, and the thermosetting resin is not limited if it can be used for the resin transfer molding. In particular, according to the present invention, as long as a matrix resin having inferior toughness is used, a composite material having excellent interlaminar debonding property is obtained, and therefore, various matrix resins can be adopted for the resin transfer molding.

Furthermore, since the preform for composite material having laminated structure according to the present invention is molded using the resin transfer molding, a

fiber-reinforced composite material having high strength and excellent interlaminar debonding property can be obtained.

### Examples

5           The present invention will be explained with reference to examples and comparative examples as follows.

Evaluation items and measuring methods for the fiber-reinforced composite material obtained by examples and comparative examples are described below.

#### 10   [Interlaminar debonding Property]

4.9 kg of an iron weight whose tip had a radius of 7.94 mm and a hemispheric shape was dropped on a composite material which was molded into flat shape. The area of interlaminar debonding (damaged area) generated by the above dropping of the iron weight was determined by a transmission method using an ultrasonic damage scanner. The area was evaluated by comparing with an area of interlaminar debonding for the composite material prepared without interlaminar reinforcement by the thermoplastic layers (reference example). Impact tests by dropping the iron weight on the composite material were carried out under the following conditions.

Condition 1: The iron weight was dropped in free-fall from a height of 32 cm.

Condition 2: The iron weight was dropped in free-fall from a height of 64 cm.

#### [Measurement of Compressive Strength After Impact (CAI)]

The iron weight was dropped at the center of a rectangular test plate 4 inches in width and 6 inches in length under the above condition 2, and further, the CAI was measured in accordance with the SACMA recommended measuring method (SRM2).

#### [Reference Example]

As a reinforcement fiber fabric, a fabric of 145 g/m<sup>2</sup> (weight per square meter) made of carbon fiber "MR50K4.5M" (trade name; manufactured by Mitsubishi Rayon Co., Ltd.) weaved by 5-harness satin weave, was used. The reinforcement fiber fabrics were laminated with a structure of lamination of [(0/90)/(±45)]<sub>8</sub>S in a mold, the mold was closed, a modified epoxy resin "#985" (trade name; manufactured by Mitsubishi Rayon Co., Ltd.) was injected under pressure at 95°C into the mold through a gate (port) provided at the side

of the mold, and the modified epoxy resin #985 was hardened at 180°C for two hours. A molded material was taken from the mold and flashings were removed to obtain a test plate for evaluation.

The test plate was subjected to the above impact test, the area of damage was determined using the ultrasonic scanner, the area was determined to be 100 as a reference, and damaged areas of the following examples and the above reference were compared by subjecting to impact under the same conditions respectively to exhibit their relative value.

#### [Example 1]

As a thermoplastic resin for reinforcing spaces between the layers, a plain fabric of 10 g/m<sup>2</sup> (count of each of weft and warp: 12 threads per inch), which was formed of long fibers composed of 2.64 d/36 filament of nylon 12, was prepared. The reinforcement fiber fabric which was the same as used in the reference example and the above fiber fabric composed of the thermoplastic resin for interlaminar reinforcement were alternately laminated in the mold to form a lamination, wherein both outermost layers of the lamination were formed of the reinforcement fiber fabrics. Except for these processes, a composite material was prepared by the same processes as described in the Reference Example. The obtained composite material was evaluated by the test for interlaminar debonding. As a result, the damaged area decreased to 73% when carrying out the test under the condition 1 and 68% when carrying out the test under the condition 2.

When the cross-section of the obtained composite material was observed, it was found that the composite material was molded without voids; thus, the liquid resin was confirmed to be able to flow in the preform without being disturbed by the fiber fabric composed of the thermoplastic resin for reinforcing spaces between the layers.

#### [Example 2]

As a thermoplastic resin for reinforcing spaces between the layers, a uniaxial fabric of 10 g/m<sup>2</sup> (count of warp: 23 threads per inch; count of weft: a pair of cord fabrics per two inches), which was woven by the same nylon 12 as used in Example 1, was prepared.

Except for the above process, a composite material was prepared by the same process as described in Example 1. The obtained composite material was evaluated by the test for interlaminar debonding. As a result, the damaged area decreased to 70% when carrying out the test under the condition 1 and decreased to 65% when carrying out the test under the

condition 2.

When the cross-section of the obtained composite material was observed, it was found that the composite material was molded without voids; thus, the liquid resin was confirmed to be able to flow in the preform without being disturbed by the fiber fabric composed of the thermoplastic resin for reinforcing spaces between the layers.

[Example 3]

As a thermoplastic resin for reinforcing spaces between the layers, a non-woven fabric was used, wherein the non-woven fabric was obtained as follows: the same nylon 12 fiber as used in Example 1 was chopped into 10 mm lengths, the chopped fiber was dispersed in water; the dispersed fibers were collected on a screen to an areal weight of 10 g/m<sup>2</sup>; and the collected fibers were heat-pressed. Except for the above processes, a composite material was prepared by the same process as described in Example 1. The obtained composite material was evaluated by the test for interlaminar debonding. As a result, the damaged area decreased to 69% when carrying out the test under the condition 1 and decreased to 66% when carrying out the test under the condition 2.

When the cross-section of the obtained composite material was observed, it was found that the composite material was molded without voids; thus, the liquid resin was confirmed to be able to flow in the preform without being disturbed by the non-woven fabric composed of the thermoplastic resin for reinforcing spaces between the layers.

[Example 4]

As a thermoplastic resin for reinforcing spaces between the layers, the same nylon 12 fiber as used in Example 1 chopped into 10 mm lengths was used. The chopped fiber was dispersed on the same reinforcement fiber fabric formed of carbon fiber as used in Reference Example, so as to be 10 g/m<sup>2</sup>, the other reinforcement fiber fabric was laminated on this reinforcement fiber fabric, and this series of processes was repeated. Except for the above processes, a composite material was prepared by the same process as described in Example 1. The obtained composite material was evaluated by the test for interlaminar debonding. As a result, the damaged area decreased to 72% when carrying out the test under the condition 1 and decreased to 68% when carrying out the test under the condition 2.

When the cross-section of the obtained composite material was observed, it was found that the composite material was molded without voids; thus, the liquid resin was

confirmed to be able to flow in the preform without being disturbed by the chopped fiber composed of the thermoplastic resin for reinforcing spaces between the layers.

[Example 5]

5 As a thermoplastic resin for reinforcing spaces between the layers, a network structure of the same nylon 12 fiber as used in Example 1 was prepared. The network structure was prepared by providing the nylon 12 fiber on a silicone rubber sheet in one direction so as to be 5 g/m<sup>2</sup> or at intervals of 2.1 mm, providing another nylon 12 fiber on the provided nylon 12 fiber in the perpendicular direction so as to be at the same pitch as the above, and heat-pressing the lamination. Except for the above processes, a composite material was prepared by the same process as described in Example 1. The obtained composite material was evaluated by the test for interlaminar debonding. As a result, the damaged area decreased to 66% when carrying out the test under the condition 1 and decreased to 63% when carrying out the test under the condition 2.

When the cross-section of the obtained composite material was observed, it was found that the composite material was molded without voids; thus, the liquid resin was confirmed to be able to flow in the preform without being disturbed by the network structure composed of the thermoplastic resin for reinforcing spaces between the layers.

[Example 6]

As a thermoplastic resin for the interlaminar reinforcement, a porous film of 10 g/m<sup>2</sup>, which was obtained by making holes 3 mm in diameter at intervals in a lattice of 5 mm squares on a nylon 12 film of 13.9 g/m<sup>2</sup>, was prepared. Except that the above film was used, a composite material was prepared by the same process as described in Example 1.

25 The obtained composite material was evaluated by the test for interlaminar debonding. As a result, the damaged area decreased to 65% when carrying out the test under the condition 1 and decreased to 61% when carrying out the test under the condition 2.

When the cross-section of the obtained composite material was observed, it was found that the composite material was molded without voids; thus, the liquid resin was confirmed to be able to flow in the preform without being disturbed by the porous film composed of the thermoplastic resin for reinforcing spaces between the layers.

Each form and amount of use of the thermoplastic resins for reinforcing spaces

between the layers used in each example, and the results of interlaminar debonding tests, are shown in Table 1.

Table 1

	Form of thermoplastic resin	Weight per square meter of thermoplastic resin layer (g/m <sup>2</sup> )	Damaged area (relative value)	
			Condition 1	Condition 2
Reference Example	none	0	100	100
Example 1	plain fabric	10	73	68
Example 2	uniaxial fabric	10	70	65
Example 3	non-woven fabric	10	69	66
Example 4	chopped fiber	10	72	68
Example 5	reticulated structure	10	66	63
Example 6	porous film	10	65	61

#### [Comparative Example 1]

The same fabric as was used in the Reference Example was used as a reinforcement fiber fabric and a modified bismaleimide resin "#2010" (trade name; manufactured by Mitsubishi Rayon Co., Ltd.) was used as a matrix resin. A composite material was prepared by the steps of: providing unreacted modified bismaleimide resin flatly in a lower mold composed of a bottom plate and an outer frame having an outlet at its side; providing a preform formed by laminating the reinforcement fiber fabric on the unreacted modified bismaleimide resin; providing an upper mold which is a small slidable lid on the preform; exhausting air contained in the preform through the outlet provided at the side of the mold; heating the preform to 100°C in a vacuum so as to impregnate it with a liquid resin (unreacted modified bismaleimide resin) in the direction of thickness of the preform; heating the mold to 180°C for six hours to harden the liquid resin; and removing the composite material from the mold. Subsequently, the composite material was heated in a forced convection oven at 232°C for six hours to carry out post-curing in order to obtain a test plate for evaluation.

The test plate was subjected to an impact under Condition 2 to determine the area of



damage, and the damaged area was compared with that of each Example.

[Example 7]

As a thermoplastic resin for the interlaminar reinforcement, a plain-woven fabric  
5 having 10 g/m<sup>2</sup> (count of each weft and warp: 6 threads per inch) was prepared, wherein the  
plain-woven fabric was formed by weaving tow of bound 48 fibers of 3.96 d made of  
polyimide resin "MATRIMID 5218" (trade name; manufactured by Ciba-Geigy).

The reinforcement fiber fabric and the above thermoplastic resin fabric for the  
interlaminar reinforcement were laminated alternately on the unreacted resin in the mold to  
10 be a laminated material, the laminated material in which the upper and lower outermost  
surfaces were made of the reinforcement fiber fabric, was molded and post-cured using the  
same process as Comparative Example 1 to obtain a test plate for evaluation.

The test plate was subjected to an impact under Condition 2 to determine the area of  
damage, and the damaged area was decreased to 83% in comparison with Comparative  
Example 1.

[Example 8]

As a thermoplastic resin fabric for the interlaminar reinforcement, a plain-woven  
fabric having 20 g/m<sup>2</sup> (count of each weft and warp: 12 threads per inch), formed by  
20 weaving the same fiber as used in Example 7, was prepared. Except that the above  
plain-woven fabric was used, a test plate for evaluation was prepared by the same process as  
described in Example 7.

The test plate was subjected to an impact under Condition 2 to determine the area of  
damage, and the damaged area was decreased to 60% in comparison with Comparative  
25 Example 1.

[Example 9]

As a thermoplastic resin fabric for the interlaminar reinforcement, a plain-woven  
fabric having 30 g/m<sup>2</sup> (count of each weft and warp: 18 threads per inch), formed by  
30 weaving the same fiber as used in Example 7, was prepared. Except that the above  
plain-woven fabric was used, a test plate for evaluation was prepared by the same process as  
described in Example 7.

The test plate was subjected to an impact under Condition 2 to determine the area of

damage, and the damaged area was decreased to 41% in comparison with Comparative Example 1.

Each amount for use of the thermoplastic resin fabric for the interlaminar reinforcement used in Comparative Example 1 and Examples 7 to 9, and the damaged areas obtained by adding each an impact under Condition 2, are shown in Table 2.

Table 2

	Example 7	Example 8	Example 9	Comparative Example 1
Weight per square meter of thermoplastic resin fiber fabric (g/m <sup>2</sup> )	10	20	30	0
Damaged area under Condition 2 (relative value)	83	60	41	100

#### [Comparative Example 2]

As a reinforcement fiber fabric, a fabric of 145 g/m<sup>2</sup> (weight per square meter) made of a carbon fiber "TR50S12M" (trade name; manufactured by Mitsubishi Rayon Co., Ltd.) woven by uniaxial fabric, was used. The reinforcement fiber fabrics were laminated with a structure of lamination of [45/0/-45/90]4S in the mold. Except for the above process, the same molding as in the Reference Example was carried out to obtain a composite material.

Compressive strength after impact (CAI) which was measured after the composite material was subjected to damage by the impact test under Condition 2, was 188 MPa.

#### [Comparative Example 3]

The reinforcement fiber fabric which was the same as that used in Comparative Example 2, was laminated with a structure of lamination of [45/0/-45/90]4S in the mold. The laminated material was stitched with thread formed of carbon fiber "TR50S12M" (trade name; manufactured by Mitsubishi Rayon Co., Ltd.) with single stitching at 5 mm stitch (a pitch of 10 mm) and 5 mm intervals between rows of single stitches and the stitched laminated material was provided in the mold. Except for the above process, the same

molding process as in the Reference Example was carried out to obtain a composite material.

The area damaged by the impact test under Condition 2 was decreased to 35% in comparison with Comparative Example 2; however, the CAI was 217 MPa.

#### 5 [Example 10]

As a thermoplastic resin fabric for the interlaminar reinforcement, the same fabric as was used in Example 1 was used. The reinforcement fiber fabric used in the Comparative Example and the thermoplastic resin fabric for reinforcing between layers were laminated alternately in the mold to form a laminated material in which the upper and lower  
10 outermost surfaces were made of the reinforcement fiber fabric. Except for the above process, a test plate for evaluation was prepared by the same process as described in Comparative Example 2.

The area damaged by the impact test under Condition 2 was decreased to 63% in comparison with Comparative Example 2, and the CAI was 255 MPa.

The evaluation results of Comparative Examples 2 and 3, and Example 10 are shown in Table 3.

#### 20 [Example 11]

A sized yarn, which was prepared by impregnating 0.01 g/m of the resin used in the Reference Example in the nylon filaments used in Example 1 and the reinforcement fiber in directions perpendicular to each other, were laminated at both surfaces of the reinforcement fiber fabric which was the same as was used in Comparative Example 2 at a pitch of 12 threads per inch to prepare a sheet. The sheet was laminated with a structure of lamination  
25 of [45/0/-45/90]4S in the mold. Except for the above process, the same molding process as in Comparative Example 2 was carried out to obtain a test plate for evaluation.

The area damaged by the impact test under Condition 2 was decreased to 60% in comparison with Comparative Example 2, and the CAI was 258 MPa.

#### 30 [Example 12]

A uniaxial fabric of carbon fiber with carbon fiber at  $145 \text{ g/m}^2$  and nylon filament at  $10 \text{ g/m}^2$  was prepared by carrying out plain-weaving of the reinforcement fiber, which was the same used in Comparative Example 2, as warp and the nylon filaments, which were the

same nylon filaments used in Example 1 was used as weft (count: 24 threads per inch). The uniaxial fabric of carbon fiber was laminated with [45/0/-45/90]4S in the mold. Except for the above process, a test plate for evaluation was prepared by the same process as described in Comparative Example 2.

- 5           The area damaged by the impact test under Condition 2 was decreased to 62% in comparison with Comparative Example 2, and the CAI was 256 MPa.

The evaluation results of Comparative Examples 2 and 3, and Examples 10, 11, and 12 are shown in Table 3.

Table 3

	Ex. 10	Ex. 11	Ex. 12	Comparative Ex. 2	Comparative Ex. 3
Damaged area under Condition 2 (relative value)	63	60	62	100	35
CAI (MPa)	255	258	256	188	217

#### Industrial Applicability

The preform of the present invention can be molded by resin transfer molding to provide a composite material comprising excellent strength and interlaminar debonding resistance.

## CLAIMS

1. A preform for a composite material, comprising layers of laminated structure composed of reinforcing material formed of reinforcement fiber and a layer provided between the layers, which is made of thermoplastic resin and has spaces so that flow of liquid resin to be injected for molding the composite material is uninhibited.
2. A preform for a composite material, according to claim 1, wherein the layer, which is made of thermoplastic resin and has space so that flow of liquid resin to be injected for molding composite material is uninhibited, is a fabric formed of thermoplastic resin fiber thread.
3. A preform for a composite material, according to claim 1 or 2, wherein the layer, which is made of thermoplastic resin and has space so that flow of liquid resin to be injected for molding composite material is uninhibited, has a weight per square meter of 1 to 50 g/m<sup>2</sup>.
4. A preform for a composite material, comprising layers of laminated structure composed of reinforcing material formed of reinforcement fiber, wherein thermoplastic resin threads are adhered or woven approximately parallel to each other on a surface of the reinforcing material.
5. A preform for a composite material, according to claim 4, the thermoplastic resin thread disposed or woven to a surface of the reinforcing material has a weight per square meter of 1 to 50 g/m<sup>2</sup>.
6. A preform for a composite material, according to any one of claims 1 to 5, wherein the reinforcing material formed of reinforcement fiber is a woven fabric.
7. A preform for a composite material, according to any one of claims 1 to 6, wherein the thermoplastic resin is one selected from the group of polyamides and polyimides.
8. A fiber-reinforced composite material formed by molding the preform for a composite material according to any one of claims 1 to 7.

# Declaration, Power Of Attorney and Petition

Page 1 of 3

WE (I) the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PREFORM FOR COMPOSITE MATERIAL AND COMPOSITE MATERIAL

the specification of which

☐ is attached hereto.

☐ was filed on \_\_\_\_\_ as

Application Serial No. \_\_\_\_\_

and amended on \_\_\_\_\_.

☒ was filed as PCT international application

Number PCT/JP00/02319

on April 10, 2000,

and was amended under PCT Article 19

on \_\_\_\_\_ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
Patent 11-101617	Japan	08/04/1999	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

_____	_____
(Application Number)	(Filing Date)
_____	_____
(Application Number)	(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.	Filing Date	Status (pending, patented, abandoned)
_____	_____	_____
_____	_____	_____
_____	_____	_____

And we (I) hereby appoint: Norman F. Oblon, Reg. No. 24,618; Marvin J. Spivak, Reg. No. 24,913; C. Irvin McClelland, Reg. No. 21,124; Gregory J. Maier, Reg. No. 25,599; Arthur I. Neustadt, Reg. No. 24,854; Richard D. Kelly, Reg. No. 27,757; James D. Hamilton, Reg. No. 28,421; Eckhard H. Kuesters, Reg. No. 28,870; Robert T. Pous, Reg. No. 29,099; Charles L. Gholz, Reg. No. 26,395; Vincent J. Sunderdick, Reg. No. 29,004; William E. Beaumont, Reg. No. 30,996; Robert F. Gnuse, Reg. No. 27,295; Jean-Paul Lavalleye, Reg. No. 31,451; Stephen G. Baxter, Reg. No. 32,884; Martin M. Zoltick, Reg. No. 35,745; Robert W. Hahl, Reg. No. 33,893; Richard L. Treanor, Reg. No. 36,379; Steven P. Weihrouch, Reg. No. 32,829; John T. Goolkasian, Reg. No. 26,142; Richard L. Chinn, Reg. No. 34,305; Steven E. Lipman, Reg. No. 30,011; Carl E. Schlier, Reg. No. 34,426; James J. Kulbaski, Reg. No. 34,648; Richard A. Neifeld, Reg. No. 35,299; J. Derek Mason, Reg. No. 35,270; Surinder Sachar, Reg. No. 34,423; Christina M. Gadiano, Reg. No. 37,628; Jeffrey B. McIntyre, Reg. No. 36,867; and Paul E. Rauch, Reg. No. 38,591; our (my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P.C., whose Post Office Address is: Fourth Floor, 1755 Jefferson Davis Highway, Arlington, Virginia 22202.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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